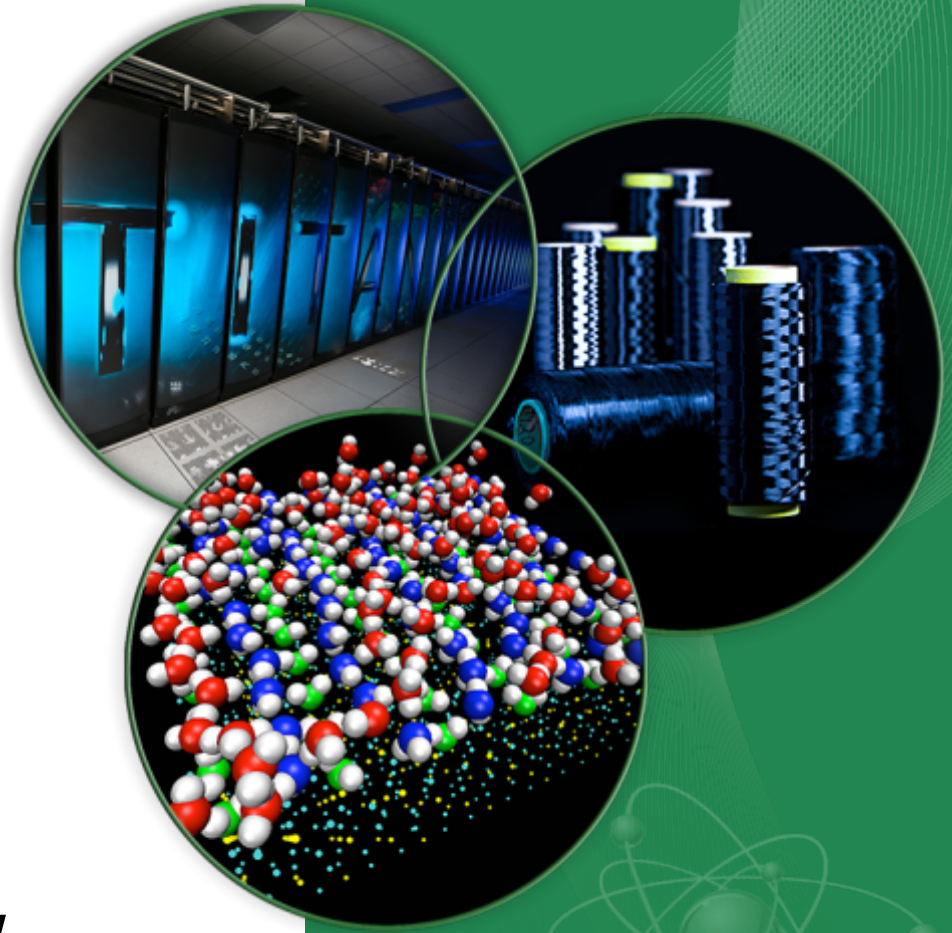


Resonance Region Cross-Section Measurements for Calcium and Cerium

Klaus Guber
**Oak Ridge National
Laboratory**
Oak Ridge, TN, USA

NCSP Technical Program Review
March 2016



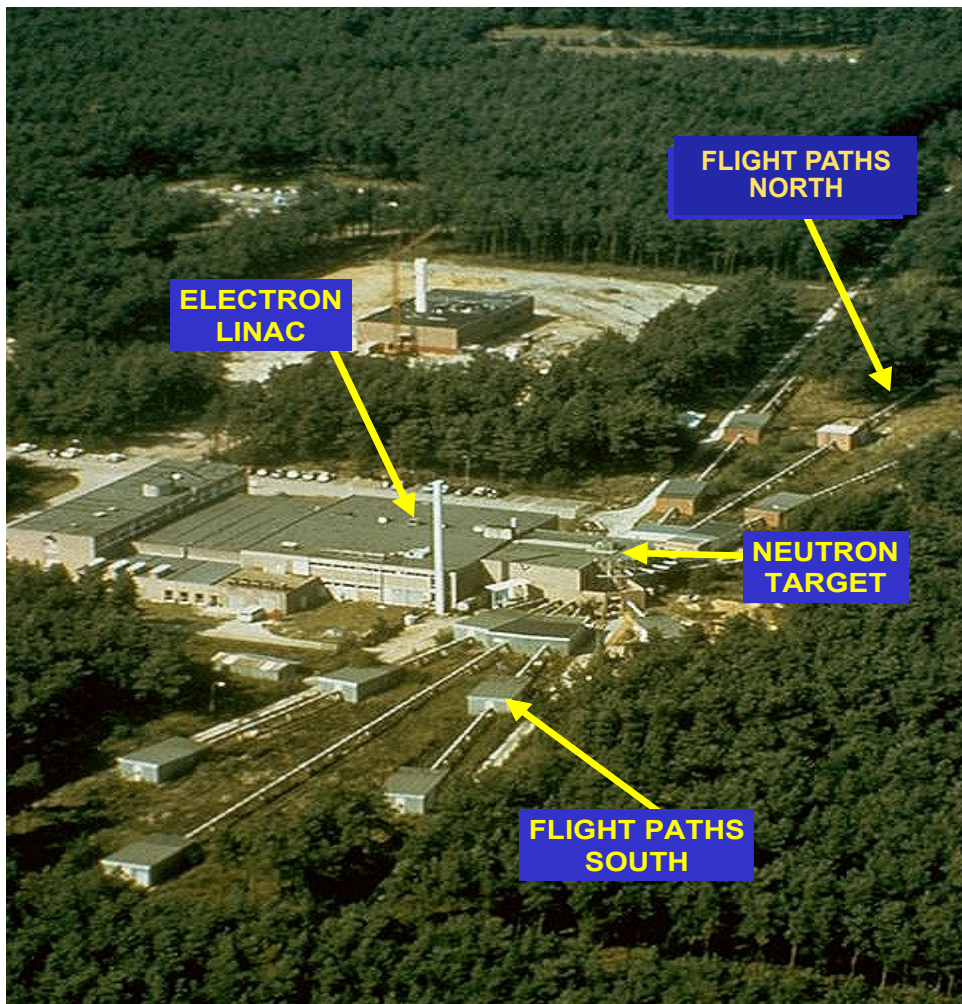
Resonance Region Nuclear Data Work for NCSP

NCSP Five Year Plan

- Objective: Provide measured and evaluated resonance-region cross-section data to address the priority NCSP nuclear data needs
- Vision: Addresses multiple Nuclear Data 5- and 10-year goals and attributes identified in the NCSP Vision
- Final product: rigorous ENDF/B resonance evaluations produced from cross-section measurements and analyses
- FY15 measurement work effort focused on calcium, cerium and vanadium—identified differential nuclear data needs by NCSP Nuclear Data Advisory Group (NDAG)

Nuclear Data										
Priority Needs */ Additional Needs			Thermal scattering (BeO, HF, D ₂ O, SiO ₂ , CH ₂ , C ₂ F ₄ , C ₅ O ₂ H ₈ , etc.), ²³⁹ Pu, Cr, ²³⁷ Np, Pb, ⁵⁵ Mn, Ti, ²⁴⁰ Pu / ²³³ U, Th, Be, ⁵¹ V, Zr, F, K, Ca, Mo, Na, La							
Completed Evaluations (FY)			Minor Actinides (13), SiO ₂ (12), ⁵⁹ Mn (12), ^{180,182,183,184,186} W (14)							
	Materials	Pre FY2015	FY2015	FY2016	FY2017	FY2018	FY2019	FY2020	FY2021	Post-FY2021
Measurements	Calcium (Ca)									
	Cerium (Ce)									
	Copper (Cu)									
	Iron (Fe)									
	Lucite (C ₅ O ₂ H ₈)									
	Tantalum (Ta)									
	Strontium (Sr)									
	Tungsten (W)									
	Vanadium (V)									
	Zirconium (Zr)									
	Polyethylene (CH ₂)									
	Materials	Pre FY2015	FY2015	FY2016	FY2017	FY2018	FY2019	FY2020	FY2021	Post-FY2021
Complete Evaluations	Calcium (Ca)									
	Cerium (Ce)									
	Cobalt (Co)									
	Copper (Cu)									
	Dysprosium (Dy)									
	Gadolinium (Gd)									
	Iron (Fe)									
	Lead (Pb)									
	Nickel (Ni)									
	Oxygen (O)									
	Rhodium (Rh)									
	Plutonium-239									
	Tantalum (Ta)									
	Strontium (Sr)									
	Tungsten (W)									
	Uranium-235									
	Uranium-238									
	Vanadium (V)									
	Zirconium (Zr)									
	Hydrofluoric Acid									
	Lucite (C ₅ O ₂ H ₈)									
	Polyethylene (CH ₂)									
		ORNL		RPI		LANL		LLNL/NCSP		
<ul style="list-style-type: none"> Requests for additional IE measurements: Ni, Mo, Cr (Fe-Cr alloys), Mn in intermediate energy range (VNIITF, NCERC). Request for measurements and evaluation of angular distributions at high energy for Cu. Continuing need for thermal scattering data. 										

*Note: work has been completed for some priority needs (e.g., ⁵⁵Mn, Ti, and Cr), and these isotopes/nucleides are maintained on the list for reference. Furthermore, the table represents the list of materials that can be addressed during the next five years under the current budget target. The additional priority needs will be addressed beyond the next five years.



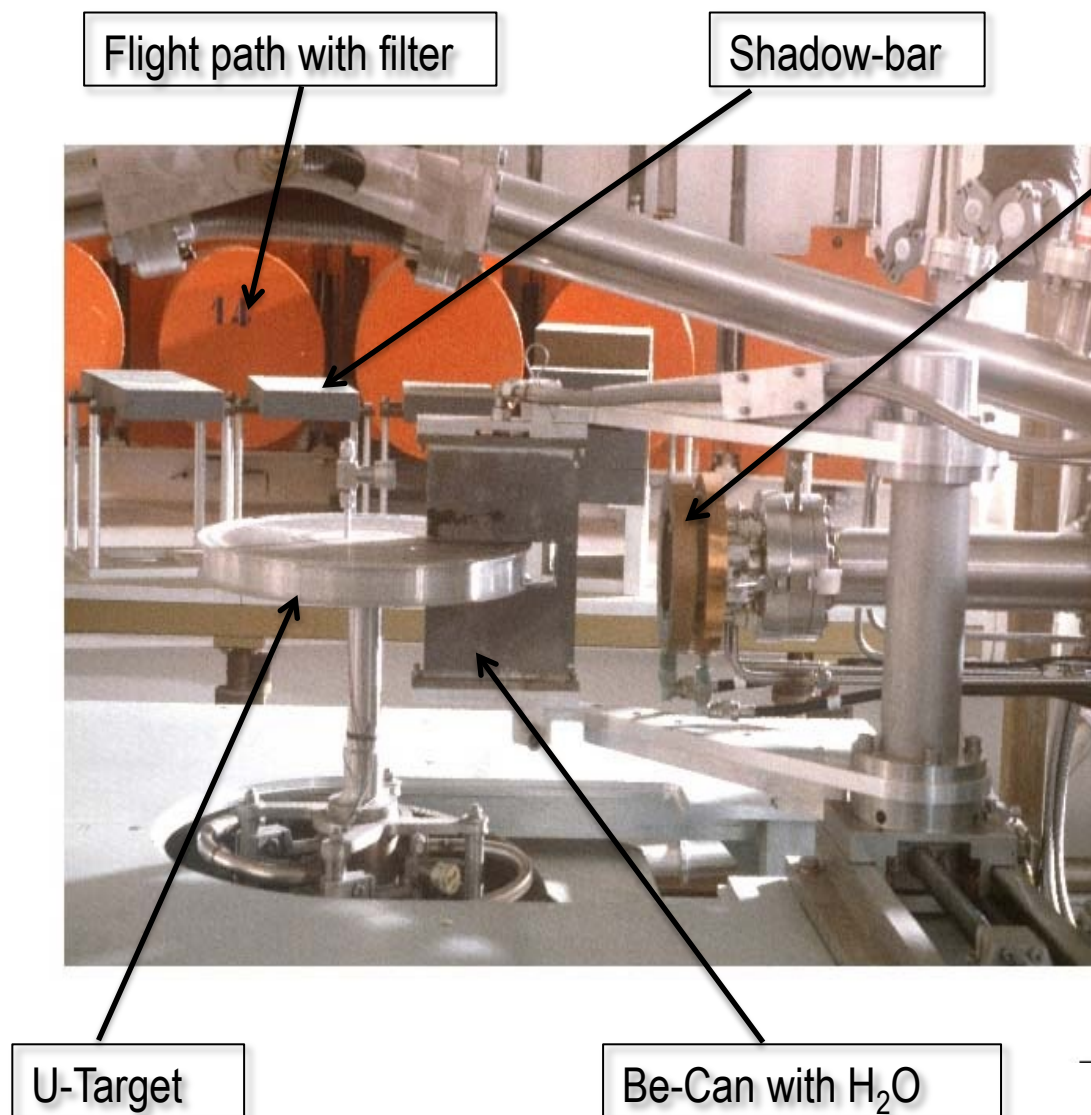
GELINA



- Time-of-flight facility
- Pulsed white neutron source
($10 \text{ meV} < E_n < 20 \text{ MeV}$)
- Multi-user facility with 10 flight paths (10 m - 400 m)
- The measurement stations have special equipment to perform:
 - Total cross section measurements
 - Partial cross section measurements

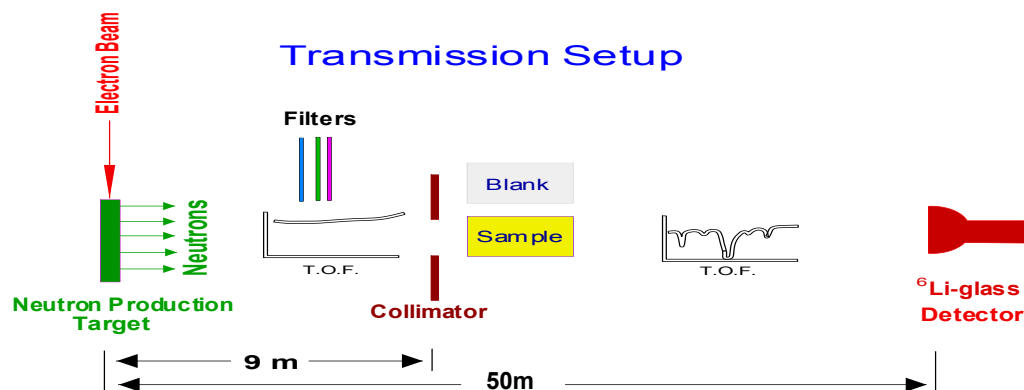
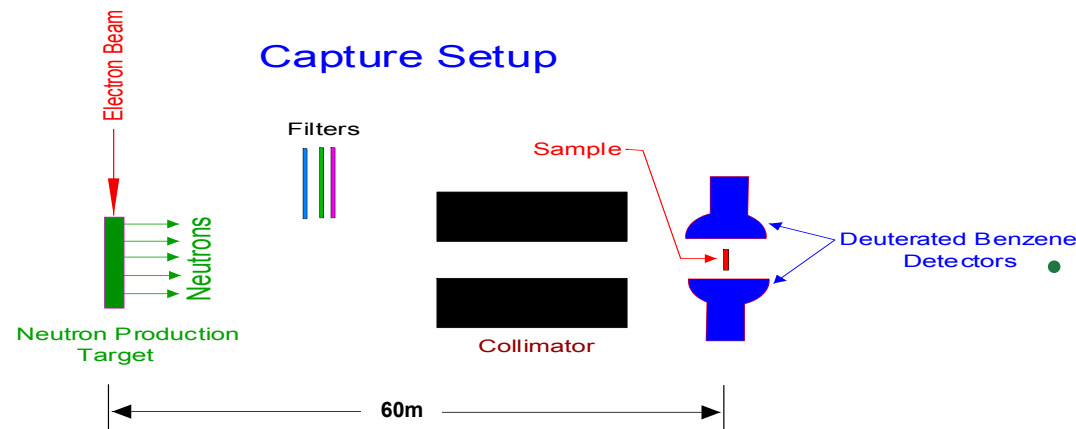
Pulse Width : 1ns
 Frequency : 40 Hz – 800 Hz
 Average Current : $4.7 \mu\text{A}$ – $75 \mu\text{A}$
 Neutron intensity : $1.6 \cdot 10^{12}$ – $2.5 \cdot 10^{13}$ n/s

Neutron Production



- e⁻ accelerated to $E_{e^-, \max} \approx 140 \text{ MeV}$
- (e⁻, γ) Bremsstrahlung in U-target (rotating & cooled with liquid Hg)
- (γ , n) , (γ , f) in U-target
- Low energy neutrons by water moderator in Be-canning

Neutron Capture and Total Cross Section Experiments at a White Neutron Source



- Time-of-flight technique is used to determine incident neutron energy. “Clocks” used have typically 1nsec resolution.

Pulsed electron beam of the accelerator starts the clock. γ -ray or neutron detector stops the clock.

$$v_n = L/t$$

$$E_n = m_n v^2/2$$

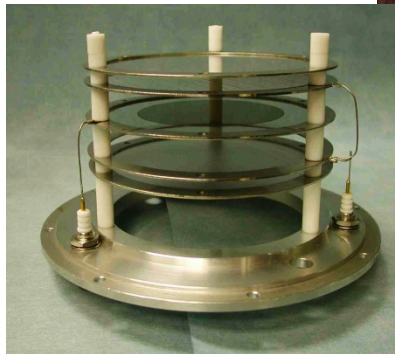
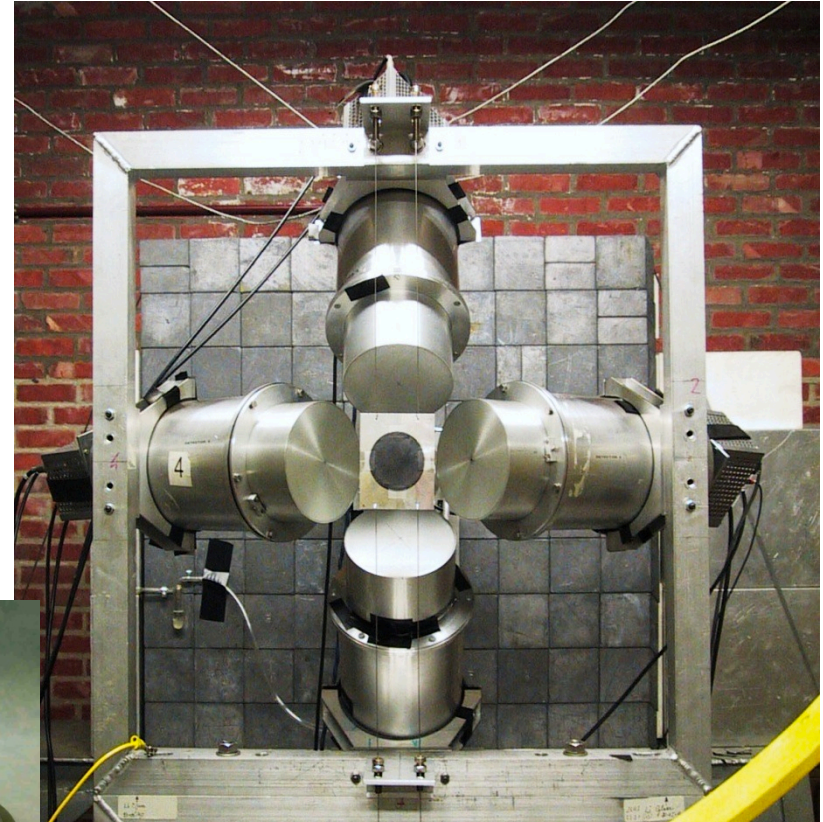
- Filters are used to reduce frame-overlap background from low-energy neutrons, reduce γ -flash effects and determine background.

Capture Cross-Section Measurements at GELINA

L = 10 m, 30 m and 60 m

Total energy detection

- C_6D_6 liquid scintillators
 - 125°
 - PHWT
 - WF from MC simulations
- Flux measurements (IC)
 - $^{10}B(n,\alpha)$
 - $^{235}U(n,f)$



$$Y_{\text{exp}} = N \sigma_{\varphi} \frac{C_w - B_w}{C_{\varphi} - B_{\varphi}}$$

Transmission Measurements at GELINA

Sample & Background Filters

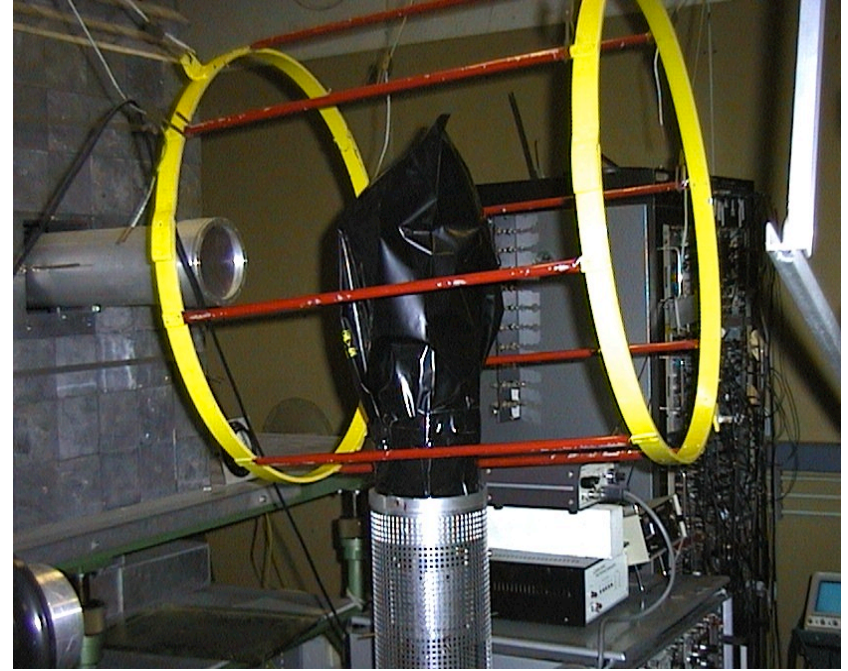


Detector stations

Moderated: L= 30 m, 50 m, (100 m, 200 m)

Fast: L= 400 m

Detector



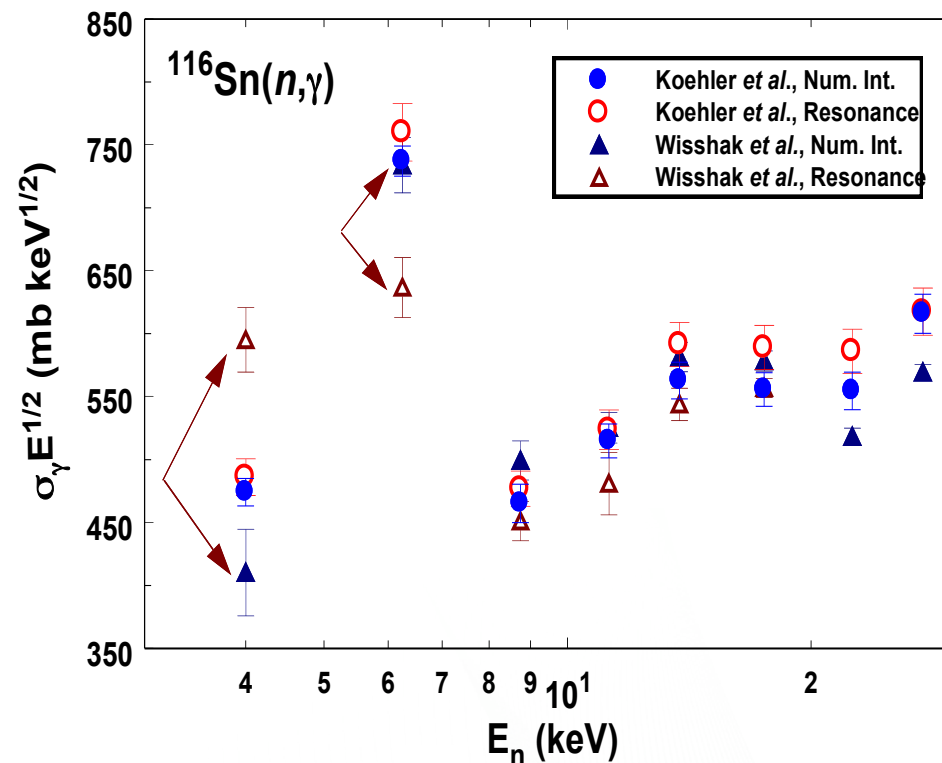
Low energy : ${}^6\text{Li}(n,t)\alpha$ Li-glass

High energy : $\text{H}(n,n)\text{H}$ Plastic scintillator

$$T = \frac{C_{in}}{C_{out}} \cong e^{-n\sigma_{tot}}$$

The Importance of Total Cross-Section Data

- More complete resonance parameter data will help improve nuclear statistical model calculations.
- Is indispensable for obtaining the most accurate (n,γ) reaction rates.
 - See resonances not very visible in (n,γ) data.
 - Since experiments are not performed with infinite small samples corrections for self-shielding and multiple scattering are needed.
- Lack of good total cross section data can lead to serious errors in these corrections and hence in the (n,γ) cross sections.



Ex: ^{116}Sn Use of incorrect neutron widths led to incorrect low-energy (n,γ) cross sections (Wisshak *et al.* PRC 54, 2732 (1996))

Thin <--> Thick transmission help to determine the statistical weight factor g

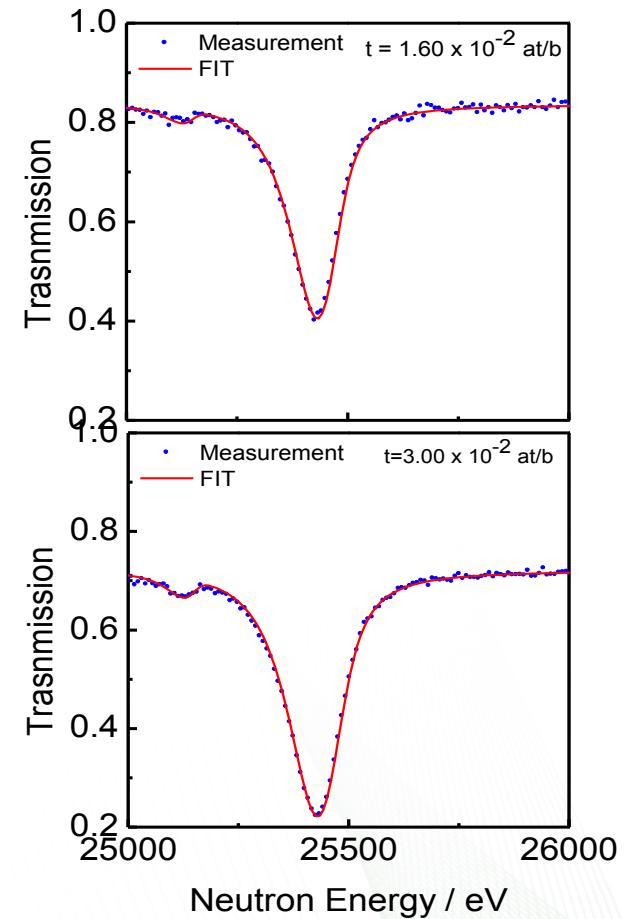
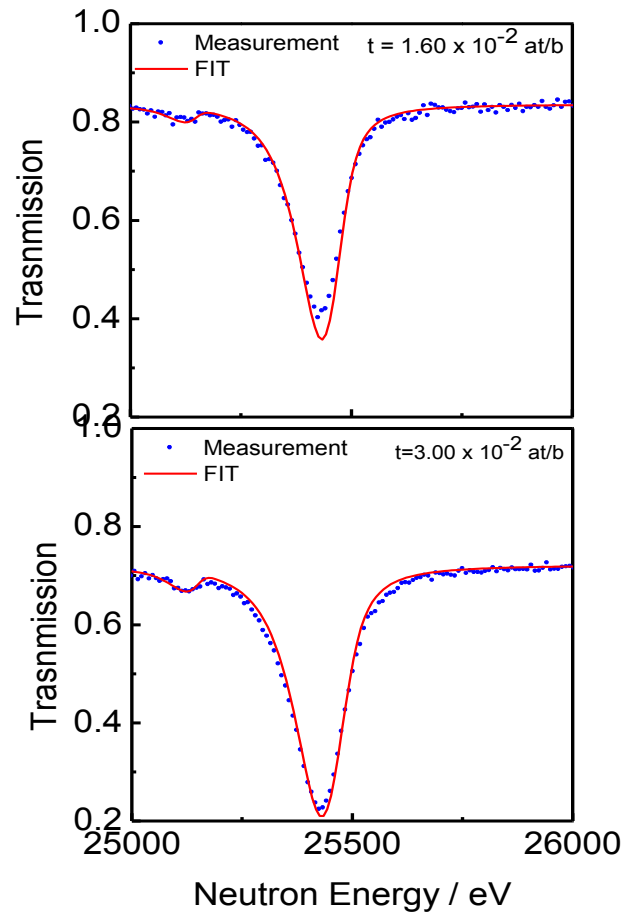
$$g = 2 \quad \frac{\chi^2}{\nu} = 2.25$$

$$g = 1 \quad \frac{\chi^2}{\nu} = 0.95$$

$$A_{t,thin} \propto ng\Gamma_n$$

$$A_{t,thick} \propto \sqrt{ng\Gamma_n\Gamma}$$

$$g = \frac{2J+1}{2(2I+1)}$$



P. Schillebeeckx, IRMM

Capture and transmission are not always complementary

$$\Gamma_{\gamma} \ll \Gamma_n$$

$$\Gamma_{\gamma} \gg \Gamma_n$$

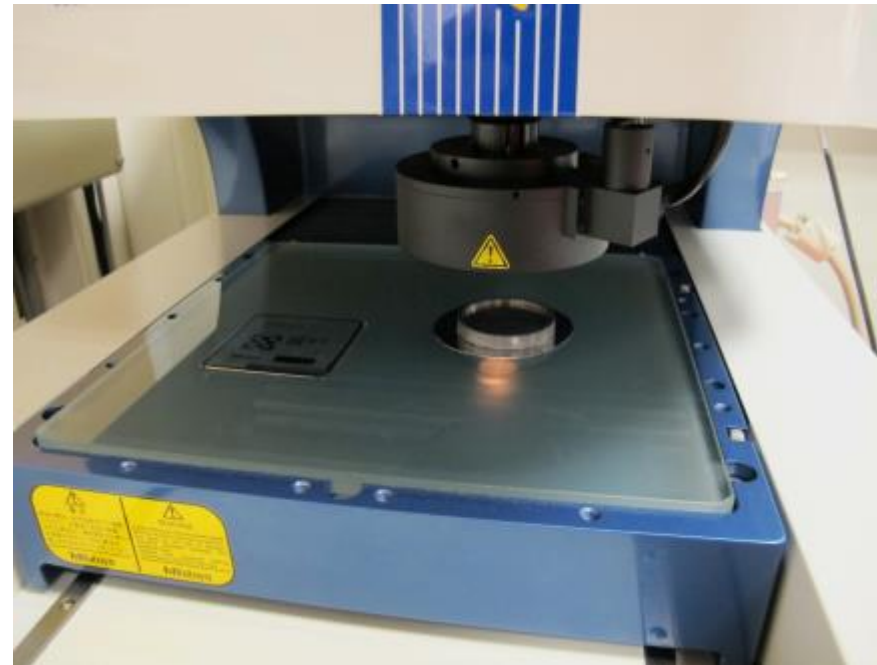
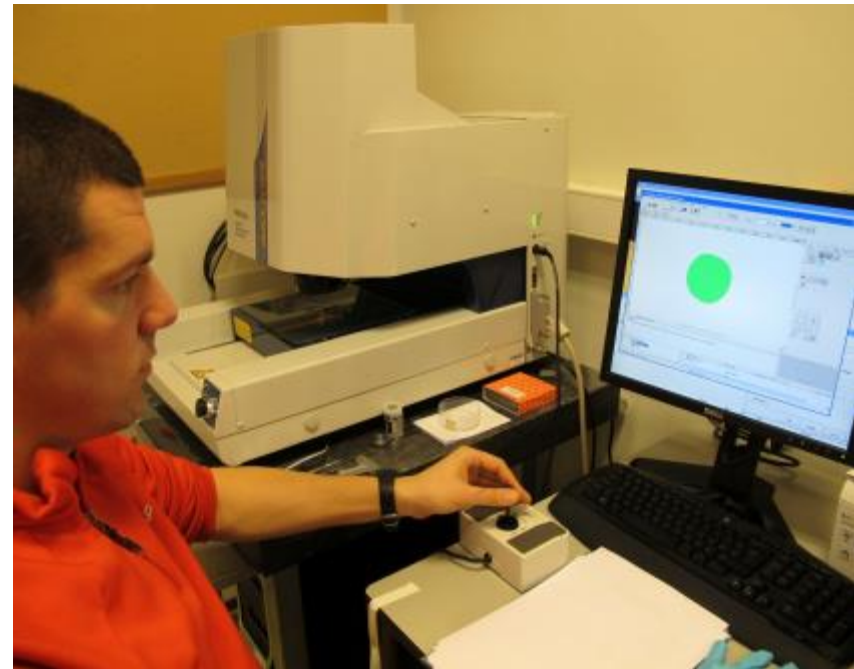
- Capture (thin): $A_{\gamma} \propto ng \frac{\Gamma_n \Gamma_{\gamma}}{\Gamma}$ $\propto ng \underline{\Gamma_{\gamma}}$ $\propto ng \underline{\Gamma_n}$
- Transmission (thin): $A_{t,thin}$ $\propto ng \underline{\Gamma_n}$ $\propto ng \underline{\Gamma_n}$

complementary

combine capture and transmission measurements with different sample thicknesses

Determine Sample Characteristics

- How much material is needed to achieve sufficient count rate.
- What material and how much is available.
- Metallic samples are preferable. See Ca. Inventory form for separated isotopes is usually oxide or carbonate.
- Physical dimensions like radius, thickness, area.
- Weight, density.
- Isotopic composition.
- Number of atoms.



Sample related problems

- Poorly characterized samples, i.e. water or impurities in the sample.
- Oxide or carbonate is inventory from of most enriched isotopes.
- Example Sm_2O_3 : Oxides are hygroscopic. There is no water free oxide!
- Even material packed and shipped under inert atmosphere contains water. Up to 30%.
- Stoichiometry not known. Is it Sm_2O_3 or $\text{Sm}(\text{OH})_3$?
- **Metallic sample are preferable**



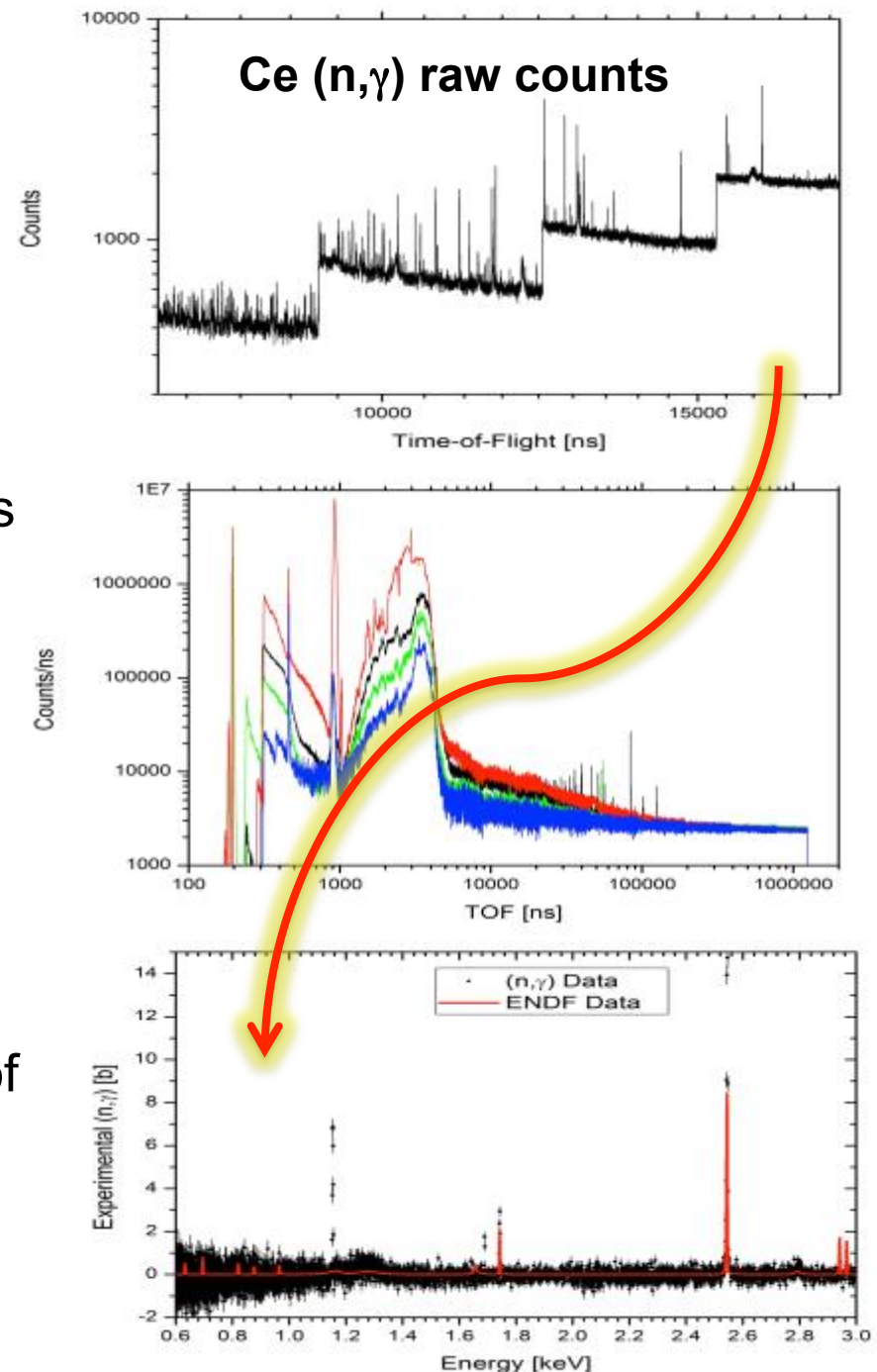
Data Taking

- Data are taken in list mode: TOF and pulse-height for detector(s) and flux monitor.
- Usually at least 2 experiments are performed for each isotope. For example, 4 isotopes translate to at least 8 experiments over the time frame of a couple of weeks each, depending on the nucleus. Different sample thickness add even more.
- Presence at GELINA is required to perform and control experiments.



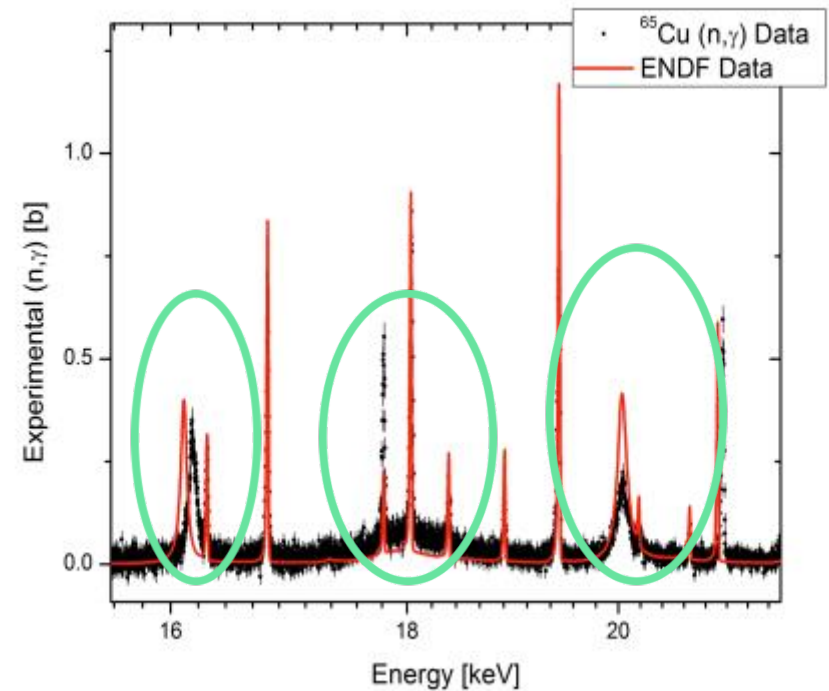
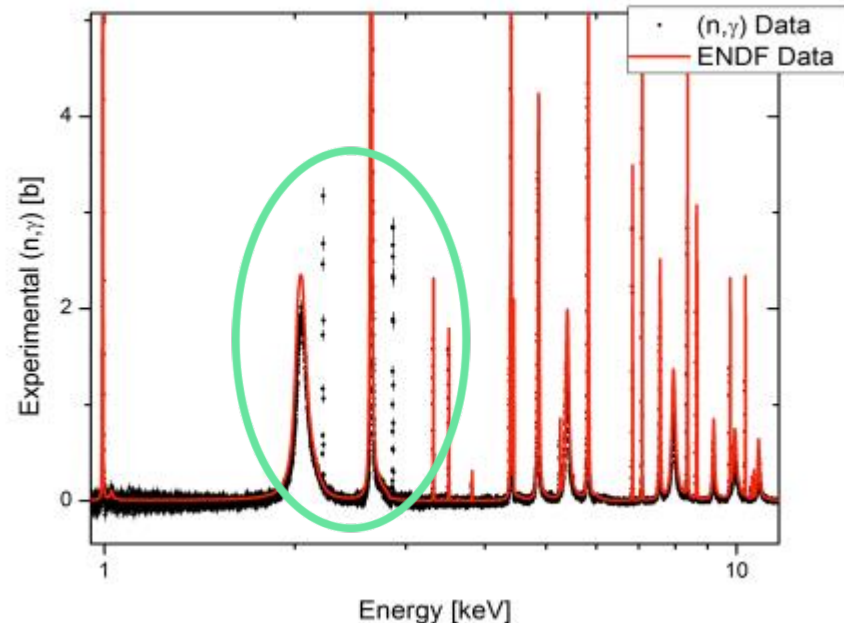
Data Reduction

- Codes for data reduction are IRMM specific and installed on IRMM computers.
- Data are sorted into spectra using the program package AGL (Analyze Geel List mode data). Raw data and scalers are tested for stability and consistency before sorting into final spectra (time-of-flight versus counts).
- The resulting spectra are then converted to cross section or transmission applying all corrections and normalizations using the AGS (Analyze Geel Spectra) code. The code is capable of a full propagation of the uncertainties for all spectra corrections and variables. A covariance matrix is generated.
- AGS installed on ORNL computers



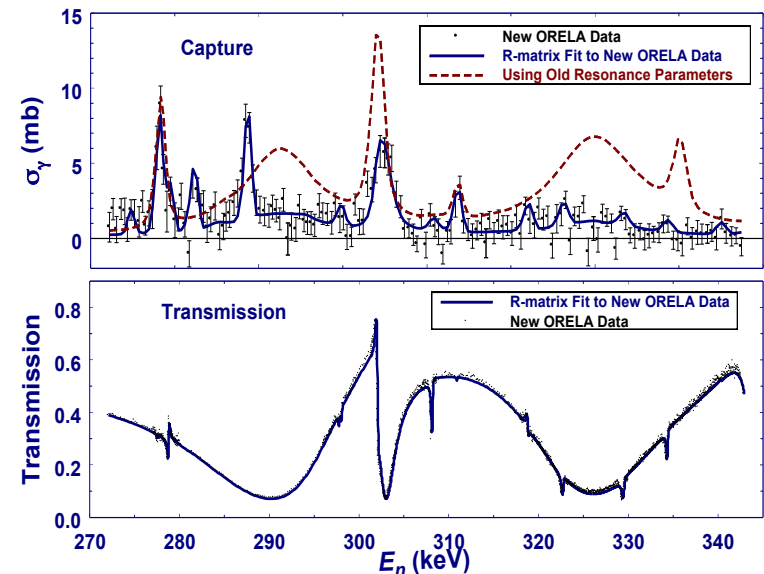
Data Testing

- Obtain all experimental information, like pulse width, repetition rate, neutron filters, flight path length, crunch table settings, sample composition and dimensions.
- Retrieve resonance parameter file for each isotope from NNDC.
- Prepare input files for SAMMY, which have to include all experimental and facility specific effects.
- For example: sample characteristics, like dimensions for multiple scattering corrections, isotopic composition, correction for applying the PHWT, resolution function, FP length, pulse width...
- Run SAMMY to check data.

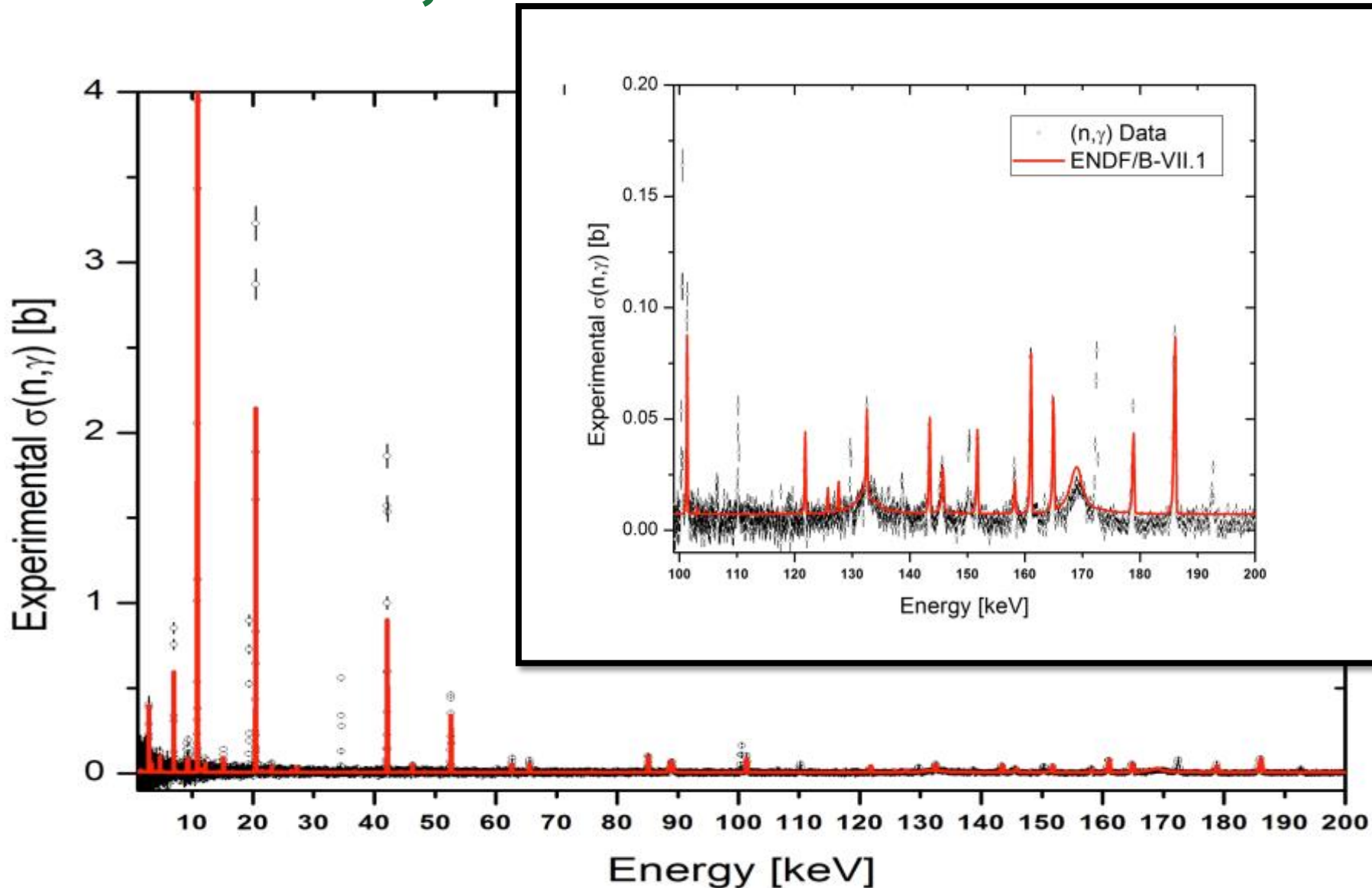


ORNL Measurement Activities for Calcium

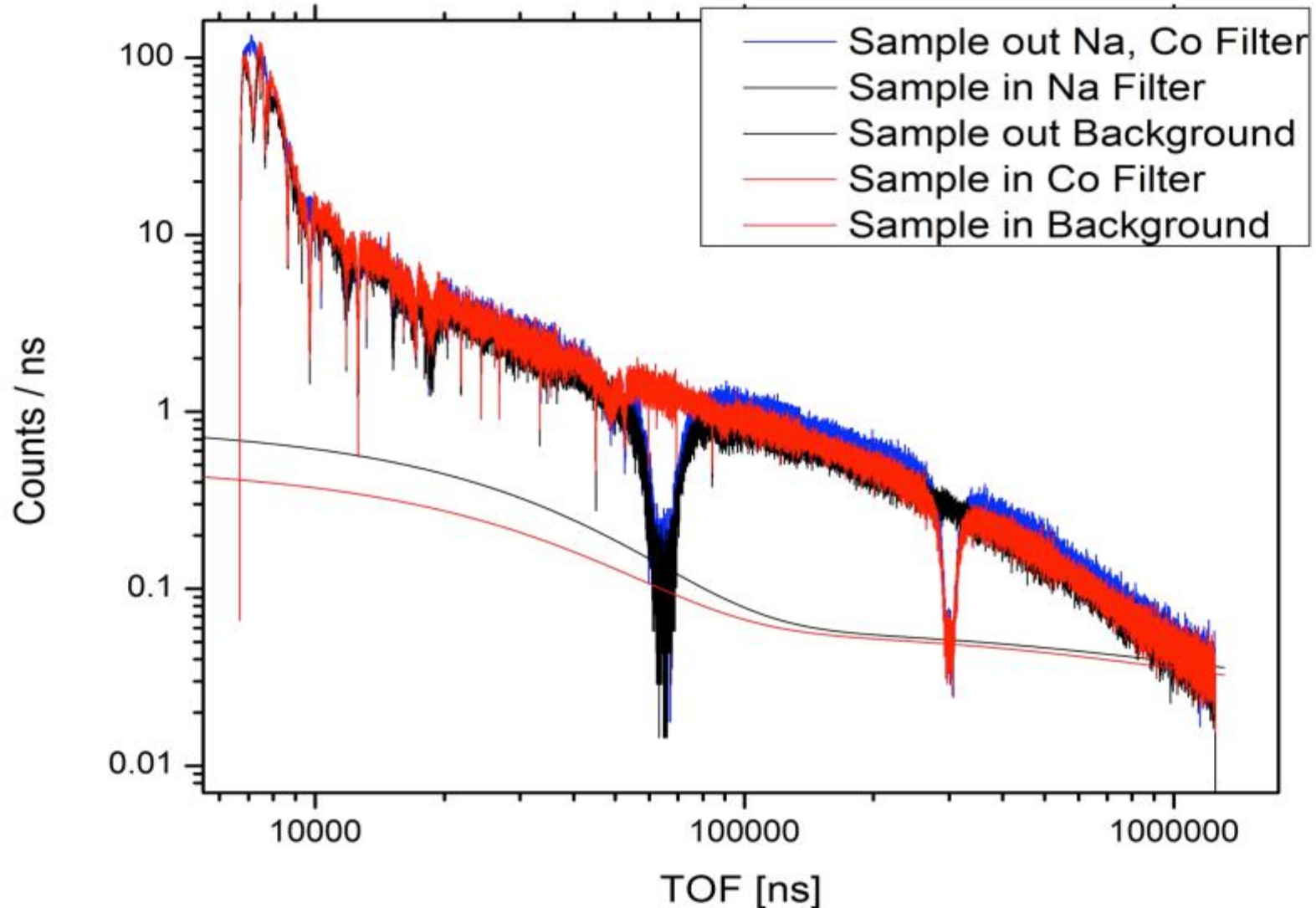
- New measurements of Ca were using metallic samples. Old (n,γ) experiments used Ca-carbonate samples (CaCO_3).
- CaCO_3 produce unwanted background in (n,γ) from sample scattered neutrons. For each Ca nucleus you have 4 nuclei which scatter only neutrons ($\text{C}+3\text{O}$).
- This background is troublesome and can produce false signal which comes within the width of the resonance of interest. Very difficult to correct for. e.g. ^{88}Sr resonances at 290, 325 keV have $\Gamma_n/\Gamma_\gamma = 18000$ and 20000
- Metallic samples increase sensitivity to small resonance in transmission.
- The samples are in thin walled Al canning due to reactivity with air.



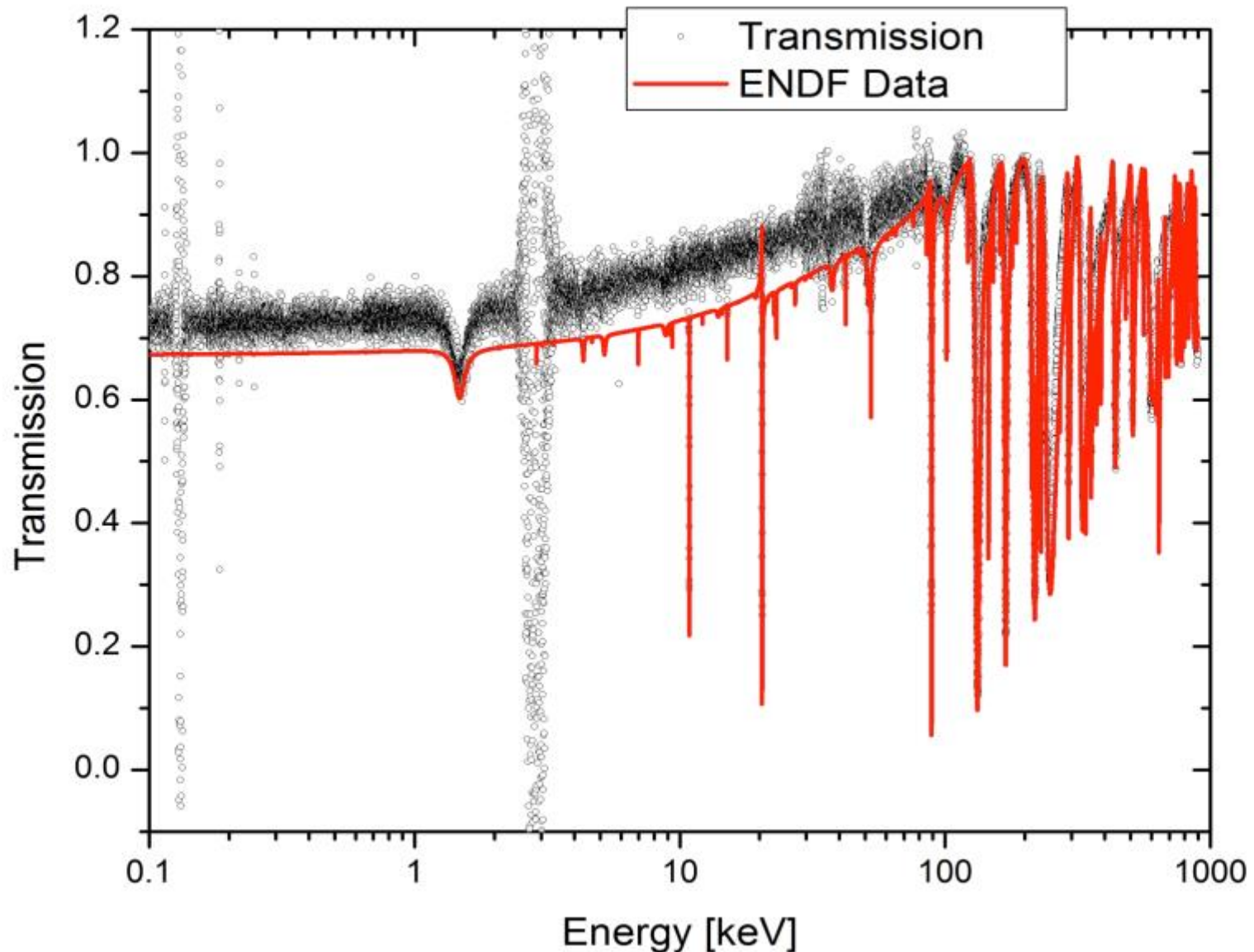
Natural Ca neutron capture using detector system at FP14, 60 m



Background Determination in Transmission using Black Resonance Filters




FP4 50m Natural Calcium Transmission of Thick Sample Compared to ENDF/B-VII



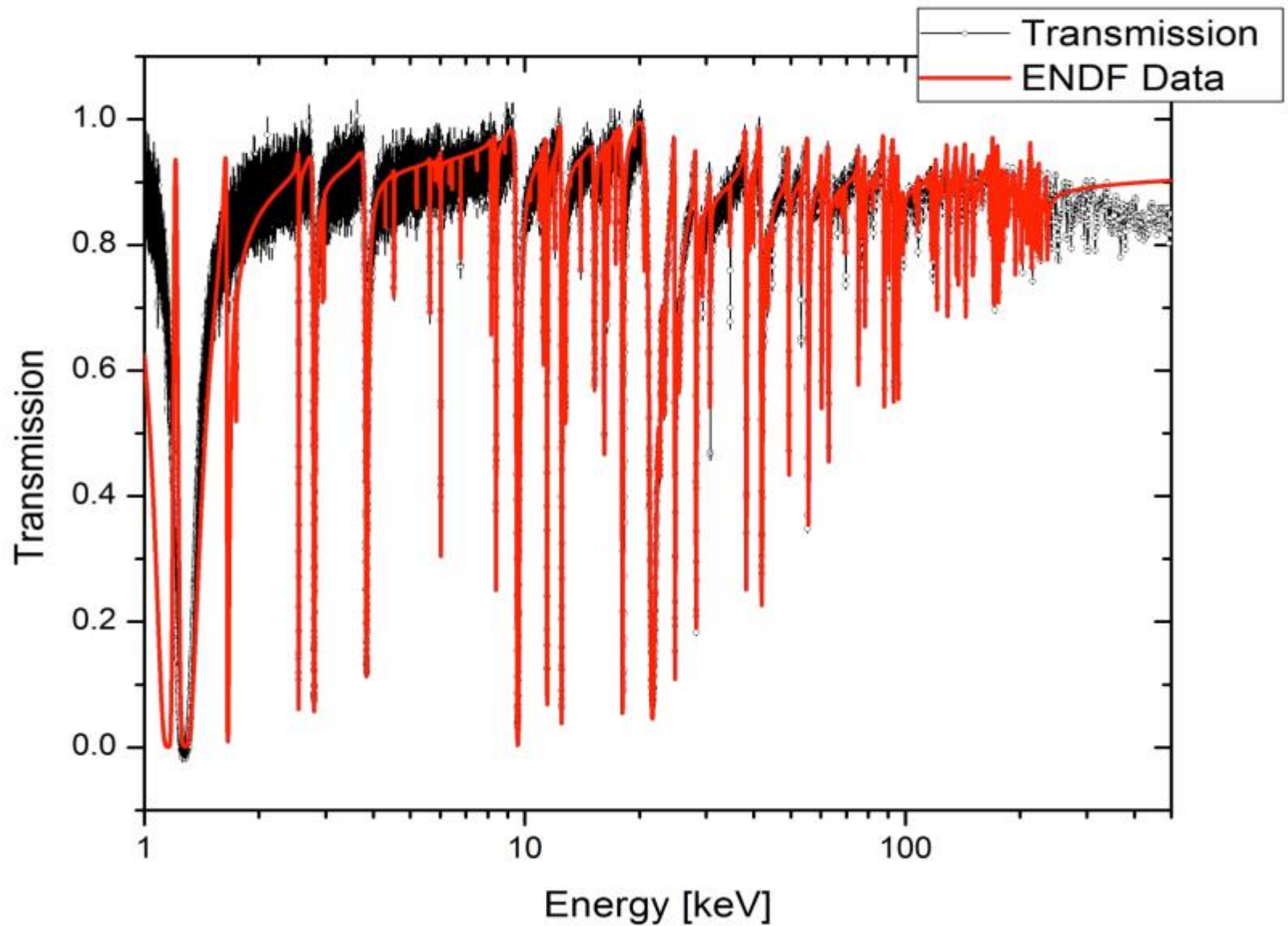
Observations

- ENDF/B-VII show for the (n,γ) data serious discrepancies compared to our data and literature (Musgrove, Nucl.Phys. A 258, 365 (1976)).
- Resonances are misassigned or missing.
- Analysis indicate to a higher capture cross section.
Reasons:
 - Pulse height weighting function method is more reliable now.
 - Older experiments suffered from background due to sample scattered neutrons. This background was often over- or underestimated.

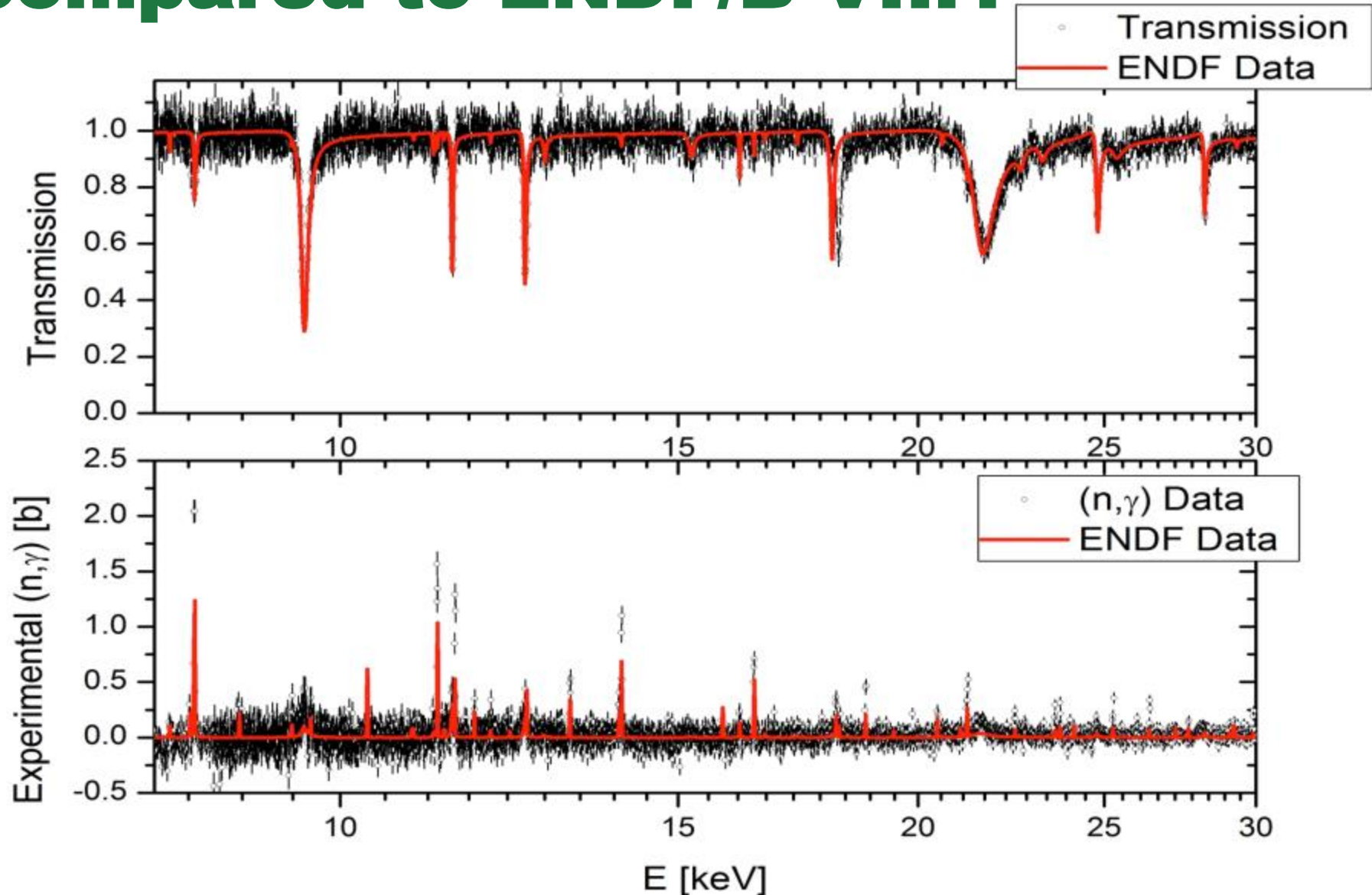
ORNL Measurement Activities for Cerium

- Measurements of Ce using natural metallic samples.
 - The samples are in Al canning due to reactivity with air.
 - Old experiments used CeCO_3 samples. Remember Ca samples.
- 
- Transmission experiments with different sample thickness were performed using FP4 50 m station.
 - Neutron capture using detector system at FP14, 60 m.
 - Experiments performed with different background filter combinations.
 - Resolving resonances above 200 keV.
 - Transmission and capture experiments using enriched Ce142 oxide sample are planned. Problems with DOE new lease policy.

Ce Transmission compared to ENDF/B-VII.1

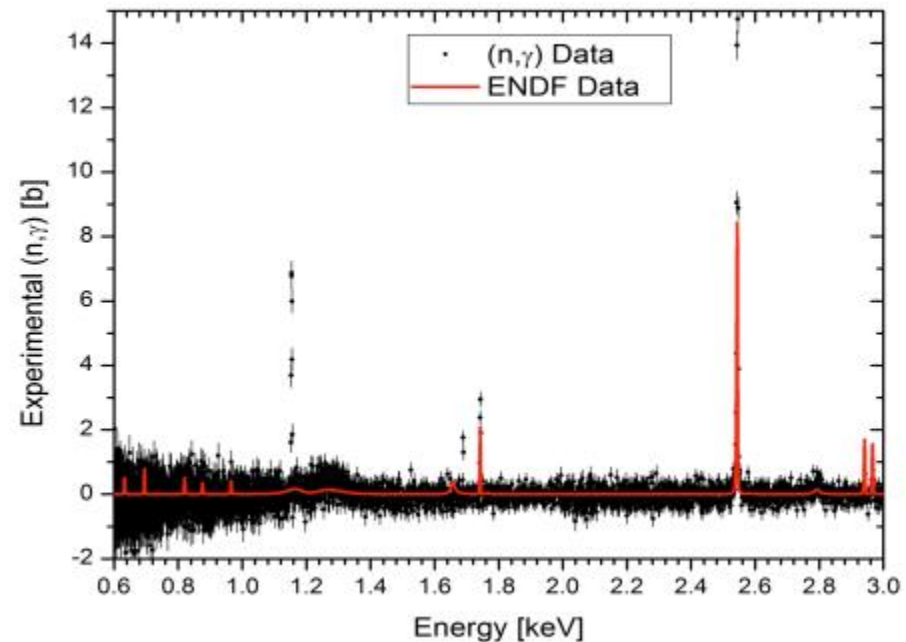
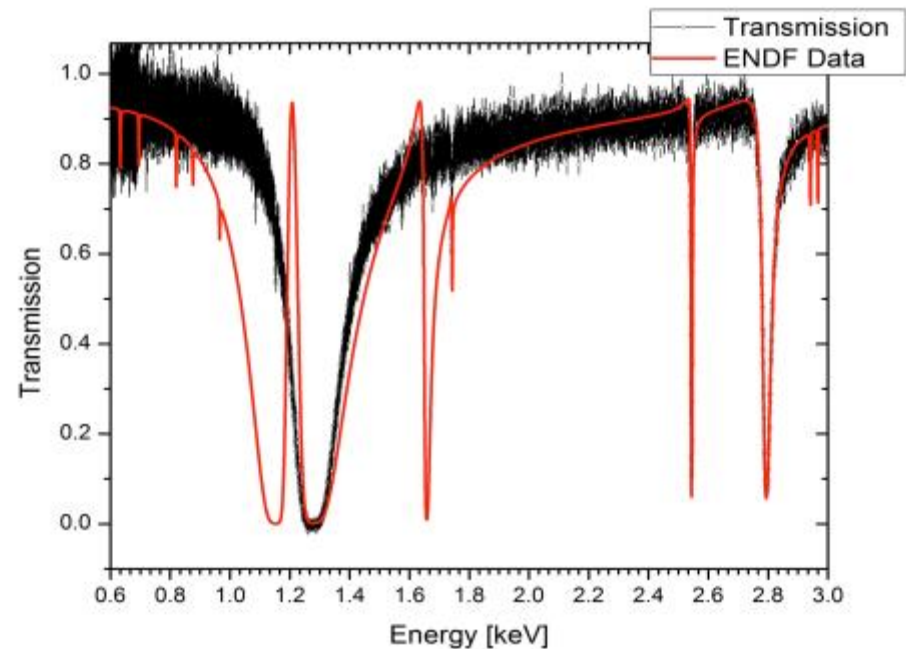


Ce Transmission and (n,γ) Data compared to ENDF/B-VII.1



Observations

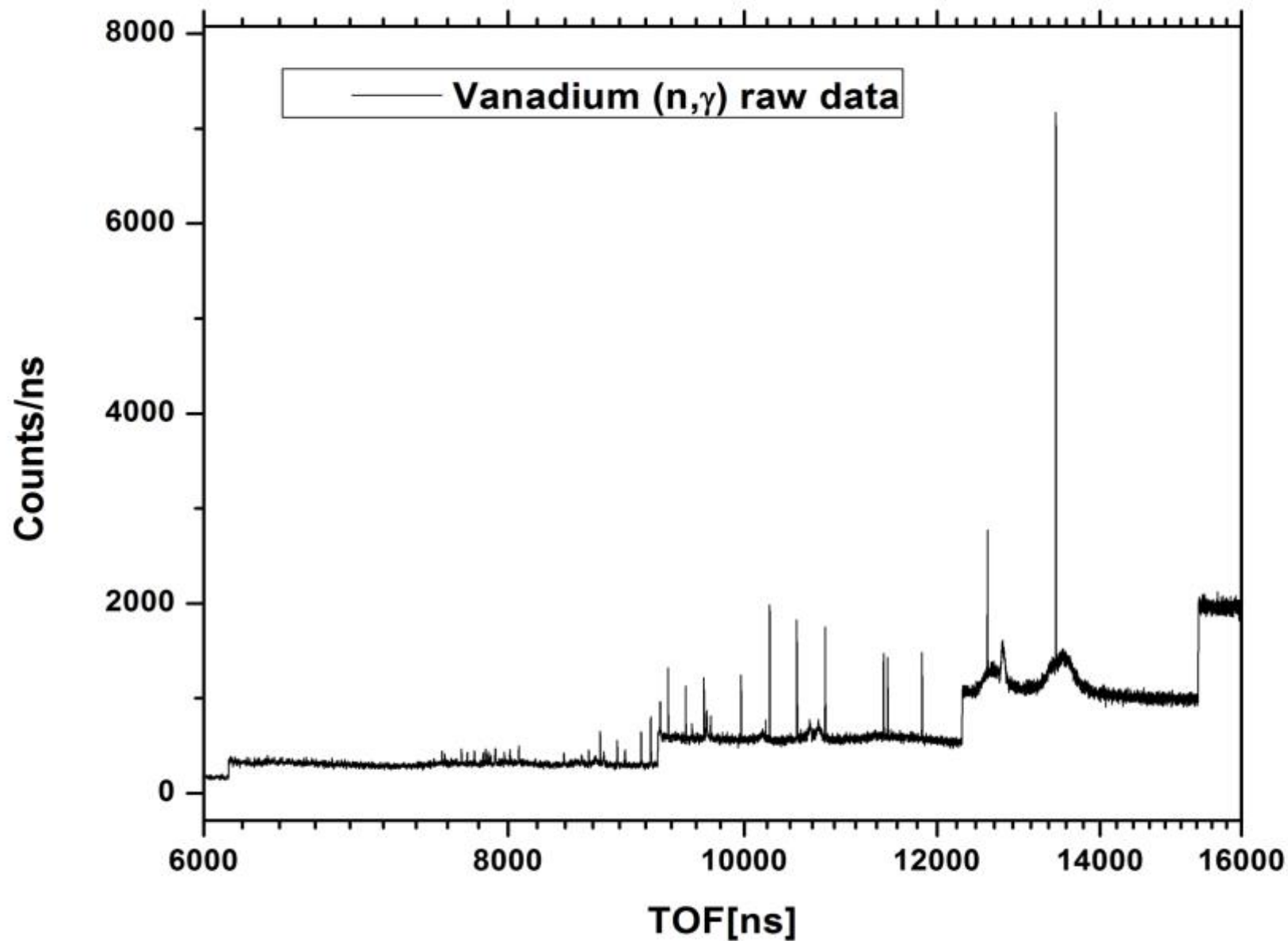
- ENDF/B-VII shows for the (n,γ) and (n,tot) data serious discrepancies compared to our data and literature.
- Resonances are misassigned or missing.
- Investigation indicate that after the reevaluation of WPEC SG23 (fission products) those resonances made it into ENDF/B VII.1
- Sometimes a reevaluation without new data and consulting the literature does not seem to be a good idea.



ORNL Measurement Activities for Vanadium

- ^{51}V 99.75% natural abundance. V is non reactive in air.
- Measurements using metallic samples of different thickness.
- Transmission experiments with different samples are performed using FP4, 50 m station.
- Neutron capture at FP14, 60 m.
- Experiments performed with different background filter combinations.
- Capture and transmission experiments performed in FY15. Need to finalize.

V (n,γ) Raw Data for Thin Sample



Status of NCSP Experiments at IRMM Geel

	W	Cu	Ca	Ce	V
Sample	metallic disks 182,183,184,186	metallic disks 63 and 65	metallic disks nat Ca	metallic disks Nat Ce, Ce-142	metallic disks
Experiments GELINA	60m, 30m (n, γ) transmission	60m (n, γ)	60m (n, γ) transmission	Nat Ce 60m (n, γ) Nat Ce transmission ¹⁴² Ce sample problems	60m (n, γ) transmission
Data Sorting	finished 60m + transmission	finished 60m	finished 60m transmission	finished for thin and thick sample	
Reduced to Cross section	X-section, transmission	X-section	X-section transmission 0.6, 1.0, 5 cm samples	2mm X-section 2mm transmission 10mm transmission	
Data Testing	Data ready for evaluation	Data ready for evaluation	Data ready for evaluation	In progress	
Analysis and Evaluation	Finalized Submitted to NNDC	Finalized Submitted to NNDC	Finalizing	Started	

People Involved in the Experiments

- Peter Schillebeeckx, IRMM
- Carlos Paradela, IRMM
- Stefan Kopecky, IRMM
- Peter Siegler, IRMM
- Ruud Wynats, IRMM
- Clint Ausmus, ORNL

People Involved in the Evaluations

- Marco Pigni, ORNL
- Vlad Sobes, ORNL
- Luiz Leal, ORNL/IRSN